IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of :
Shigetoshi SASANO et al.
Serial No. 10/055,928
Filed: January 28, 2002
For: Laminate Adhesive

Group Art Unit 1711
Examiner: Mr. Rabon A. Sergent

Declaration Under Rule 132

Honorable Commissioner of Patent and Trademarks, Washington, D.C.

Sir:

I, Shigetoshi SASANO declare:

That I am a citizen of Japan, residing at 1-20, Rikyumaemachi 1-chome, Suma-ku, Kobe, Japan;

That I was born on September 14, 1964 in Shiga and graduated from the Department of Applied Chemistry, Faculty of Engineering, Grad-school of Kobe University, Japan in March 1989;

That I have been employed by Takeda Chemical Industries, Ltd., Japan, since April 1989, and have gone on loan to MITSUI TAKEDA CHEMICALS, INC. since April 2001, and have been engaged in the research in the field of Polyurethane Adhesives;

That I am the first named inventor of U.S. Patent Application Serial No. 10/055,928; and

That the following experiments were conducted by myself according to the Example 1 set forth in the specification of US 3,892, 820;

Experiments

1. Preparation of an adhesive solution

Sixty parts of polyethylenepropylene adipate (A) (ethylene glycol/propylene glycol = 70/30 by part) having an average molecular weight of 1,200 was heated to 100℃ in an atmosphere of N_2 gas, and dehydrated and dried for 30minutes under a reduced pressure of 3 mm Hg. Then 18 parts of xylylene diisocyanate (D) (meta-isomer/para-isomer = 80/20 by part) was added to the dried (A), and allowed to react at 100℃ for 60 minutes with stirring, in the atmosphere of N_2 gas. Thus an isocyanate-terminated polyurethane prepolymer derived from said adipate (A) and xylylene diisocyanate (D) was obtained.

Separately, 69 parts of a polyester oligomer (B) having an average molecular weight of 1,780, OH-value of 62.7, COOH value of 0.3, and a melting point of 90℃-100℃ and 22 parts of a polyamide oligomer (C) having an average molecular weight of 2,000, NH_2 content of 9.75 eq./10^4g, COOH content of 0.24 eq./10^2g, and a melting point of 142℃., were together dissolved in 113 parts of dehydrated dimethylacetamide by heating in a nitrogen atmosphere. The oligomer (B) was composed of 15 mol% of naphthalene-2,6-dicarboxylic acid, 65 mol % of terephthalic acid, 20 mol % of isophthalic acid as the acid component reacted with ethylene glycol. Also the oligomer (C) was composed of 65 parts of 6,6-nylon and 35 parts of 6,10-nylon. To the solution 0.2g of dibutyltin dilaurate was added as the catalyst, and further the first-mentioned polyurethane prepolymer was added under vigorous stirring at 140℃. The reaction was thus performed for 60 minutes at approximately 140℃.

In the above-described embodiment, the composition of the reaction of the reaction system was as follows.

i. (D)/(A)+(B)+(C)(mol ratio) = 0.96
ii.(B) + (C)/(A)(weight ratio) = 1.52
iii.(C)/(B)+(C)(weight ratio) = 0.24
iv.(C)/(A)+(B)+(C)+(D)(weight %) = 13.0

In this preparation, 65 parts of 6,6-nylon was used for preparation of the

oligomer (C) instead of 30 parts of 6-nylon and 35 parts of 6,6-nylon discribed in the Example 1 of US 3,892,820, because synthesis of 6-nylon having an average molecular weight of 2,000 had been failed.

2. Evaluation

The adhesive solution thus produced was prepared for the use as a laminate adhesive. Using the prepared laminate adhesive, a composite film was produced in a method as mentioned below.

3. Production of Composite Film

A three-layered composite film of a polyethylene terephthalate film (12 μm in thickness)/an aluminum foil (9 μm in thickness)/an unextended polypropylene film (70 μm in thickness, as subjected to corona discharge treatment) was produced by the following method.

The laminate adhesive was applied onto an aluminum surface of the two-layered composite film made by adhesive bonding the polyethylene terephthalate film and the aluminum foil in advance. The spread of the adhesive was 2.5g/m^2 by weight of a solid content of the adhesive per unit area. Then, the surface applied with the adhesive was adhesive bonded with the unextended polypropylene film. Thereafter, the bonded films were cured at 50℃ for 3 days, for the curing of the adhesive.

4. Elution Test

A bag was made from the composite films thus produced and then was filled with ion-exchange distilled water as the content by the amount of 0.5mL/cm^2 per unit area of the interior surface of the bag. Then, the bag was sterilized by heated water under the pressure of 19.6×10^4 Pa at 120℃ for 30 minutes. During the sterilization, delamination of the bag made from the composite film occurred. Therefore, the elution test could not be conducted.

5. Results

The delamination was observed during the sterilization of the bag made from the composite film. Therefore, the laminate adhesive thus produced according to the Example 1 of USP 3,892,820 could not maintain adhesive

property for lamination, when the bag made from the composite film adhesive bonded with the laminate adhesive thus produced was sterilized by heated water under the pressure.

I, the undersigned, declare that all statements made herein on my knowledge are true and that all statements made on information and belief are believed to be true: and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and such willful false statements may jeopardize the validity of the application or any issuing thereon.

Signed this 30TH day of July, 2004.

Shigetoshi Sasano

Shigetoshi SASANO